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Radioactivity in the Risø District July-December 2015

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DTU Nutech Report

Sven P. Nielsen, Kasper G. Andersson and Arne Miller
DTU-Nutech-13(EN)
June 2016

DTU Nutech
Center for Nuclear Technologies

Author: Sven P. Nielsen, Kasper G. Andersson and Arne Miller
Title: Radioactivity in the Risø District July-December 2015
Center for Nuclear Technologies

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Abstract (max. 2000 char.): The environmental surveillance of the Risø environment was continued in July-December 2015. The mean concentrations in air were: 0.24±0.18 μBq m^{-3} of ^{137}Cs, 2.71±0.88 mBq m^{-3} of ^{7}Be and 0.28±0.20 mBq m^{-3} of ^{210}Pb (±1 S.D.). The depositions by precipitation at Risø in the second half of 2015 were: 0.085±0.011 Bq m^{-2} of ^{137}Cs, 607±30 Bq m^{-2} of ^{7}Be, 59.6±4.2 Bq m^{-2} of ^{210}Pb and 1.4±0.2 kBq m^{-2} of ^{3}H. The average background dose rate (TLD) at Risø (Zone I) was measured as 42 nSv h^{-1} compared with 40 ± 2 nSv h^{-1} (±1 S.D.) in the four zones around Risø.

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INTRODUCTION

A specific monitoring programme in the vicinity of the nuclear installations at the Risø site is carried out by DTU Nutech on behalf of and as a contractor to Danish Decommissioning (DD). This report presents the analytical results of the monitoring and sampling carried out in the period July-December 2015. The materials and methods used in connection with the monitoring programme are described in pages 23-24.

Table 1. Radionuclides in ground level air collected at Risø (cf. Figs. 1, 1.1 and 1.2), July-December 2015 (Unit: $\mu Bq\ m^{-3}$)

Date		^{7}Be	^{137}Cs	^{210}Pb
29-Jun-14 –	06-Jul-15	2632(10%)*	0.159(11%)	252(10%)
06-Jul-15 –	13-Jul-15	2343(10%)	0.083(14%)	122(10%)
13-Jul-15 –	20-Jul-15	2265(10%)	0.138(13%)	103(10%)
20-Jul-15 –	27-Jul-15	2246(10%)	0.417(12%)	97(10%)
27-Jul-15 –	03-Aug-15	3045(10%)	0.196(15%)	168(10%)
03-Aug-15 –	10-Aug-15	3429(10%)	0.328(13%)	288(10%)
10-Aug-15 –	17-Aug-15	5192(10%)	0.182(11%)	397(10%)
17-Aug-15 –	24-Aug-15	4008(10%)	0.469(10%)	434(10%)
24-Aug-15 –	31-Aug-15	2711(10%)	0.203(11%)	244(10%)
31-Aug-15 –	07-Sep-15	1754(10%)	0.176(12%)	157(10%)
07-Sep-15 –	14-Sep-15	2461(10%)	0.348(10%)	419(10%)
14-Sep-15 –	21-Sep-15	2256(10%)	0.990(13%)	240(10%)
21-Sep-15 –	28-Sep-15	1941(10%)	0.280(12%)	234(10%)
28-Sep-15 –	05-Oct-15	2888(10%)	0.200(10%)	383(10%)
05-Oct-15 –	12-Oct-15	3025(10%)	0.250(10%)	277(10%)
12-Oct-15 –	19-Oct-15	4093(10%)	0.168(10%)	669(10%)
19-Oct-15 –	26-Oct-15	2843(10%)	0.243(11%)	126(10%)
26-Oct-15 –	02-Nov-15	2750(10%)	0.140(10%)	739(10%)
02-Nov-15 –	09-Nov-15	2159(10%)	0.116(11%)	667(10%)
09-Nov-15 –	16-Nov-15	1430(10%)	0.101(12%)	35(10%)
16-Nov-15 –	23-Nov-15	1306(10%)	0.151(16%)	84(10%)
23-Nov-15 –	30-Nov-15	1263(10%)	0.246(14%)	49(10%)
30-Nov-15 –	07-Dec-15	3737(10%)	0.142(14%)	65(10%)
07-Dec-15 –	14-Dec-15	2357(10%)	0.205(25%)	197(10%)
14-Dec-15 –	21-Dec-15	2918(10%)	0.238(12%)	537(10%)
21-Dec-15 –	28-Dec-15	2099(10%)	0.066(20%)	131(10%)
Mean		2714	0.243	281
SD		877	0.184	202

∗Figures in brackets are relative standard uncertainties

Table 2.1. Radionuclides in precipitation in the 10 m^2 rain collector at Risø (cf. Fig. 8.1), July - December 2015. (Unit: Bq m^{-3})

Month	^{7}Be	^{137}Cs	^{210}Pb
July	1543(10%)*	0.164(19%)	112(10%)
August	2712(10%)	0.387(15%)	504(10%)
September	1803(10%)	0.112(15%)	160(10%)
October	2561(10%)	1.147(25%)	183(10%)
November	782(10%)	0.102(19%)	60(13%)
December	1130(10%)	0.095(25%)	64(10%)

*Figures in brackets are relative standard uncertainties

Table 2.2. Radionuclides in precipitation in the 10 m^2 rain collector at Risø (cf. Fig. 8.1), July - December 2015. (Unit: Bq m^{-2})

Month	Precipitation (m)	^{7}Be	^{137}Cs	^{210}Pb
July	0.055(10%)*	85.1(10%)	0.0090(25%)	6.2(14%)
August	0.049(10%)	134.0(10%)	0.0191(18%)	24.5(14%)
September	0.059(10%)	106.9(10%)	0.0066(29%)	9.5(14%)
October	0.021(10%)	55.1(10%)	0.0247(16%)	3.9(14%)
November	0.175(10%)	137.1(10%)	0.0179(25%)	10.5(16%)
December	0.079(10%)	88.8(10%)	0.0074(35%)	5.0(14%)
Sum	0.438(5%)	606.9(5%)	0.0847(13%)	59.6(7%)

*Figures in brackets are relative standard uncertainties

Table 2.3. Tritium in precipitation collected at Risø (cf. Figs. 1, 2.3.1 and 2.3.2). July - December 2015. (Unit: kBq m^{-3})

Month	10 m^2 rain collector*
July	2.1(31%)[a]
August	2.2(26%)
September	3.0(66%)
October	2.1(34%)
November	4.7(20%)
December	1.1(53%)
Double determinations*.	

[a] Figures in brackets are relative standard uncertainties

Table 2.4. Tritium in precipitation collected at Risø (cf. Fig. 1). July - December 2015. (Unit: kBq m^{-2})

Month	Precipitation (m)	10 m^2 rain collector
July	0.055(10%)[a]	0.116(32%)
August	0.049(10%)	0.108(28%)
September	0.059(10%)	0.177(67%)
October	0.021(10%)	0.043(35%)
November	0.175(10%)	0.823(22%)
December	0.079(10%)	0.087(54%)
Sum	0.438(5%)	1.354(17%)

[a] Figures in brackets are relative standard uncertainties

Table 3.1. Radionuclides in sediment samples collected at Bolund in Roskilde Fjord.(cf. Fig. 3.1) July - December 2015. (Unit: Bq kg^{-1} dry)

Date	^{137}Cs	K*
1 July	2.6(12%)[a]	16.9(10%)
*Unit: g kg^{-1} dry		

[a] Figures in brackets are relative standard uncertainties

Table 4.1. Radionuclides in seawater collected in Roskilde Fjord (cf. Fig. 4.1) July – December 2015. (Unit: Bq m^{-3})

The seawater sample has not yet been analysed for ^{137}Cs due to reconstruction of laboratories in autumn 2015 and spring 2016.

Table 4.2. Tritium in seawater collected in Roskilde Fjord (Risø pier) (cf. Fig. 4.2) July - December 2015.

Month	kBq m^{-3}
September	< 2.1 *
December	< 1.7 *
* Double determinations	

Table 5.1. Radionuclides in grass (snow) collected at Risø near the Waste Treatment Station, location I P3, Fig. 1, July - December 2015. (**Measured on bulked ash samples)*

Week no. or month	Date	K (g kg^{-1} fresh)	^{137}Cs (Bq kg^{-1} fresh)	^{137}Cs (Bq m^{-2})
27	29 June	2.7(10%) [a]	<0.2	
29	13 July	3.8(10%)	<0.2	
31	27 July	4.7(10%)	<0.2	
33	10 August	5.6(10%)	<0.4	
35	24 August	2.3(11%)	<0.3	
37	7 September	0.3(10%)	<0.3	
39	21 September	4.5(10%)	<0.5	
41	5 October	4.6(10%)	<0.5	
43	19 October	9.7(11%)	<0.3	
45	2 November	5.1(10%)	<0.1	
47	16 November	3.3(10%)	<0.3	
49	30 November	4.7(12%)	<1.3	
51	14 December	1.9(11%)	<0.2	
**July		8.8(10%)	0.181(18%)	0.051(21%)
**August		5.0(10%)	0.138(20%)	0.048(23%)
**September		4.2(10%)	0.135(22%)	0.047(25%)
**October		4.5(10%)	0.091(26%)	0.042(29%)
**November		3.9(10%)	0.143(19%)	0.030(22%)
**December		2.8(10%)	0.101(32%)	0.039(35%)

[a] Figures in brackets are relative standard uncertainties

Table 5.2. Radionuclides in Fucus vesiculosus collected at Bolund in Roskilde Fjord. July - December 2015. (Unit: Bq kg^{-1} dry)

Date	^{137}Cs	K*	% dry matter
1 July	2.0(13%)[a]	25(10%)	20(±2)
*Unit: g kg^{-1} dry			

[a] Figures in brackets are relative standard uncertainties

Table 7.1. Waste water collected at Risø (cf. Fig. 1), July - December 2015.

Week number	eqv. mg KCl l^{-1}	^{137}Cs (Bq m^{-3})	^{131}I (Bq m^{-3})	^{226}Ra (Bq m^{-3})
27	111.78	382(26%)[a]	<144	<312
28	123.02	281(27%)	<78	<146
29	81.17	<283	<444	<589
30	96.20	161(33%)	<79	<137
31	70.44	<92	<102	<222
32	77.75	168(47%)	<135	<309
33	83.53	163(49%)	<130	<294
34	100.24	<119	<129	<287
35	93.92	<128	<142	<324
36	82.14	<125	<139	<308
37	97.24	<131	<137	<291
38	94.35	<134	<138	<314
39	96.67	113(40%)	<75	<138
40	104.56	<113	<131	<290
41	127.32	<137	<136	<317
42	94.78	<134	<135	<316
43	106.71	<137	<141	<333
44	97.43	<125	<132	<298
45	117.76	<129	<135	<310
46	117.42	<144	<140	<321
47	97.91	<135	<153	<326
48	62.91	<119	<125	<272
49	26.21	<131	<53	<314
50	35.56	<122	<128	398(45%)
51	55.68	<119	<133	<313
52	65.21	<136	<161	<335
53	19.51	<80	<148	<144
Mean	83.9	<152	<138	<295
SD	30.0			

[a] Figures in brackets are relative standard uncertainties

Table 8.1. Background dose rates around the border of Risø (cf. Fig. 8.1) measured with thermoluminescence dosimeters (TLD) in the period May 2015 – October 2015. (Results are normalized to nSv h^{-1})

Location	nSv h^{-1} ¤
1	28(10%)[a]
2	36(10%)
3	36(10%)
4	40(10%)
5	43(10%)
6	146(10%)[b]
Mean	37(5%)

[a] Figures in brackets are relative standard uncertainties
[b] It is believed that the measurement result is correct, but it is unknown what caused the deviation from the normally observed value.

Table 8.2. Background dose rates around Risø (cf. Fig. 8.2 and Fig. 1) measured with thermoluminescence dosimeters (TLD) in the period May 2015– October 2015. (Results are normalized to nSv h^{-1}),

Risø zone	Location	nSv h^{-1} ¤
I	1	38(10%)[a]
I	2	33(10%)
I	3	63(10%)
I	4	38(10%)
I	5	41(10%)
Mean		43 (5%)
II	P1	37(10%)
II	P2	45(10%)
II	P3	33(10%)
II	P4	39(10%)
Mean		39 (5%)
III	P1	48(10%)
III	P2	39(10%)
III	P3	40(10%)
Mean		42 (6%)
IV	P1	32(10%)
IV	P2	33(10%)
IV	P3	43(10%)
IV	P4	38(10%)
IV	P5	35(10%)
IV	P6	39(10%)
IV	P7	52(10%)
Mean		39 (4%)
V	P1	305(10%)[b]
V	P2	-[c]
V	P3	52(10%)
V	P4	38(10%)
V	P5	41(10%)
V	P6	32(10%)
V	P7	38(10%)
V	P8	50(10%)
V	P9	40(10%)
V	P10	44(10%)
Mean		42 (4%)

[a] Figures in brackets are relative standard uncertainties

[b] It is believed that the measurement result is correct, but it is unknown what caused the deviation from the normally observed value.

[c] The TLD dosimeter has been lost during the exposure period

Table 8.3. Terrestrial dose rates at the Risø zones (cf. Fig. 8.2 and Fig. 1) July – December 2015. Measured with a NaI(Tl) detector. (Unit: nSv h^{-1})

Risø zone	Location	October
I	P1	43(10%)[a]
I	P2	55(10%)
I	P3	371(10%)
I	P4	48(10%)
I	P5	54(10%)
Mean		114(5%)
II	P1	48(10%)
II	P2	48(10%)
II	P3	43(10%)
II	P4	48(10%)
Mean		47(5%)
III	P1	51(10%)
III	P2	52(10%)
III	P3	48(10%)
Mean		50(6%)
IV	P1	40(10%)
IV	P2	49(10%)
IV	P3	48(10%)
IV	P4	49(10%)
IV	P5	34(10%)
IV	P6	51(10%)
IV	P7	46(10%)
Mean		45(4%)
V	P1	66(10%)
V	P2	55(10%)
V	P3	69(10%)
V	P4	53(10%)
V	P5	51(10%)
V	P6	58(10%)
V	P7	53(10%)
V	P7a	48(10%)
V	P8	54(10%)
V	P9	44(10%)
V	P10	54(10%)
Mean		43(4%)

[a] Figures in brackets are relative standard uncertainties

Fig. 1. Locations for measurements of gamma-background radiation Zone I and II (cf. Tables 8.2 and 8.3)

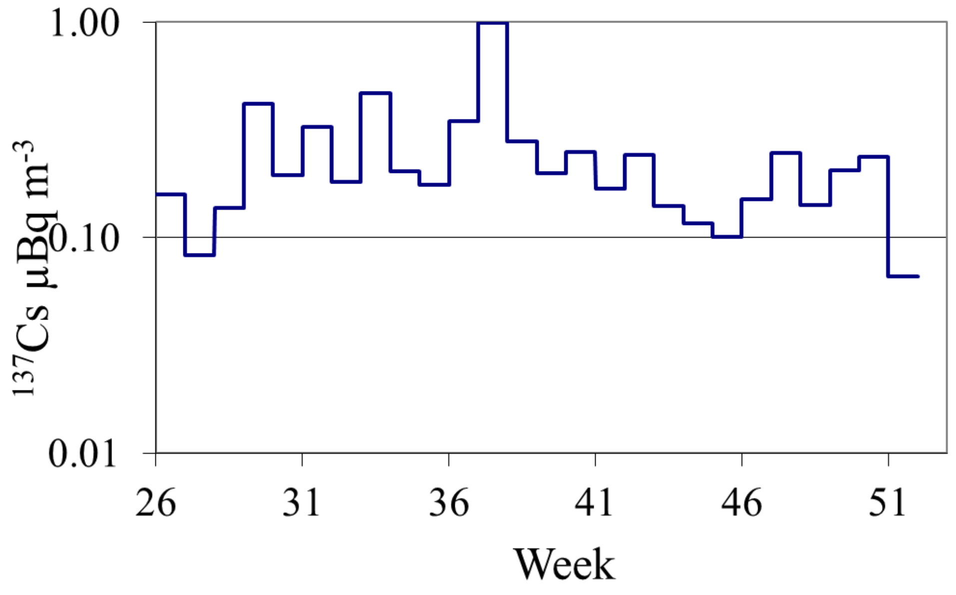

Fig. 1.1. Caesium-137 in ground level air collected at Risø in July-December 2015. (Unit: µBq m^{-3})

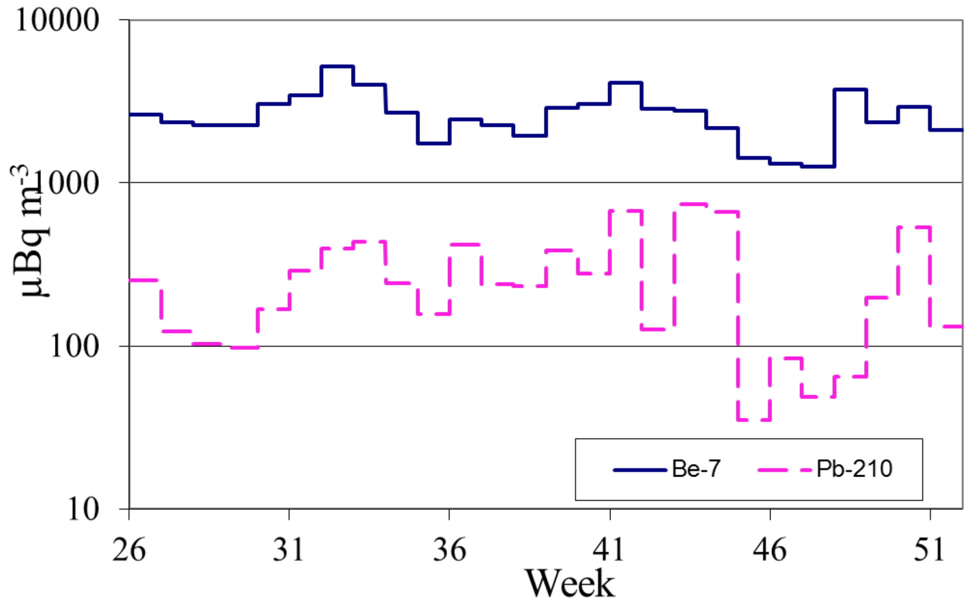

Fig. 1.2. Beryllium-7 and Lead-210 in ground level air collected at Risø in July-December 2015. (Unit: µBq m^{-3})

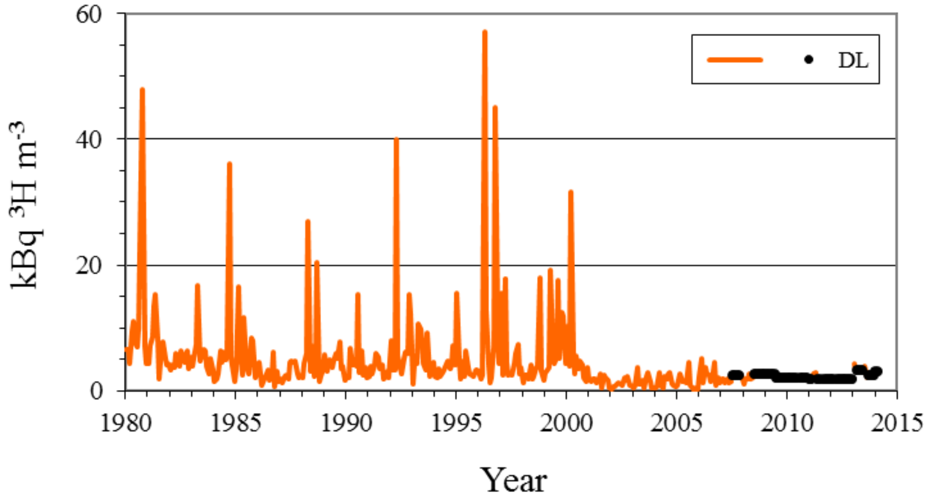

Fig. 2.3.1. Tritium in precipitation collected at Risø (1 m^2 rain collector) 1980 - 2013. (Unit: kBq m^{-3}; DL = detection limit . This rain collector was taken out of operation in 2013.

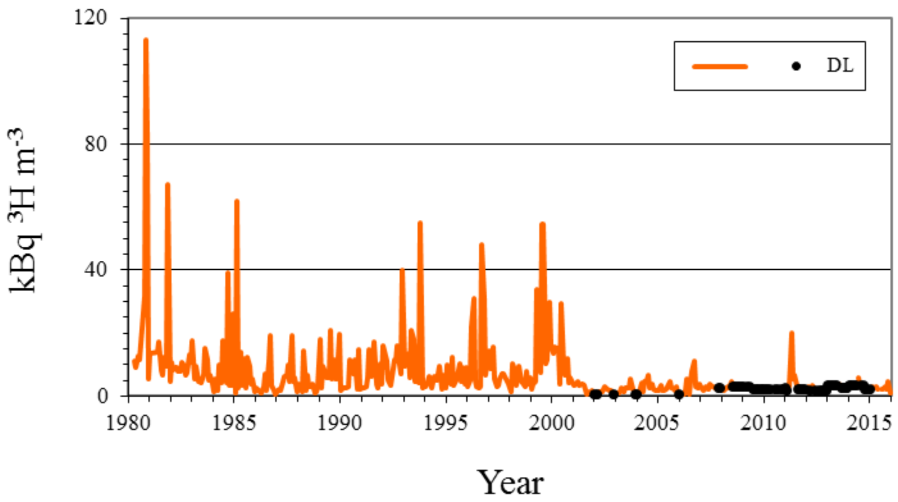

Fig. 2.3.2. Tritium in precipitation collected at Risø (10 m^2 rain collector) 1980 - 2015. (Unit: kBq m^{-3}; DL = detection limit)

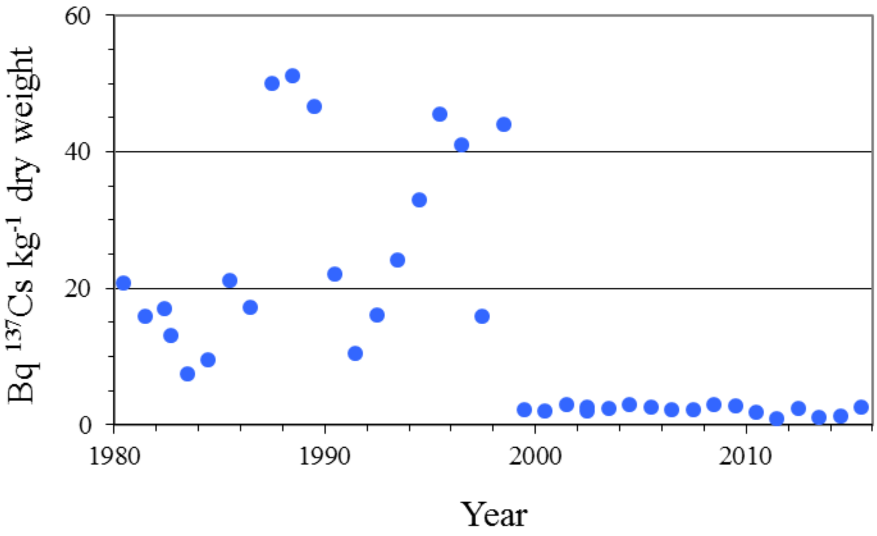

Fig. 3.1. Caesium–137 in sediment samples collected at Bolund in Roskilde Fjord. 1980 – 2015. (Unit: Bq kg^{-1} dry matter)

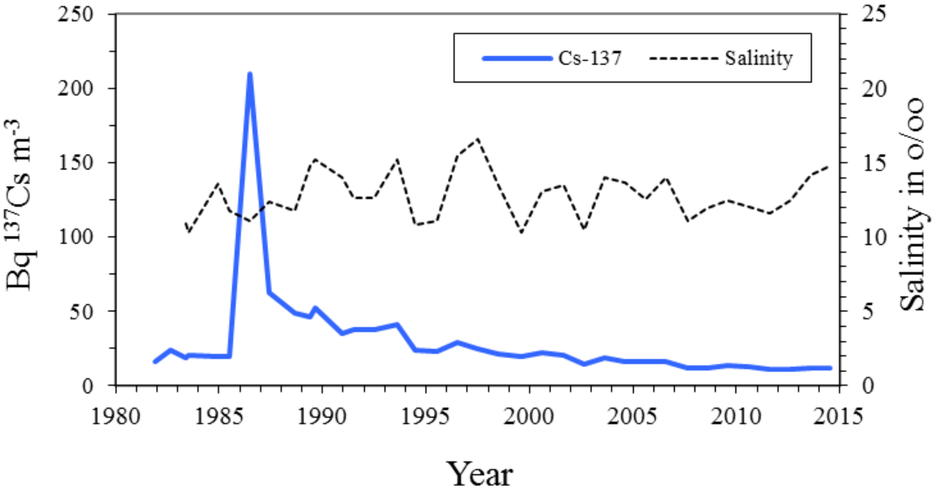

Fig. 4.1. Caesium–137 in seawater collected in Roskilde Fjord 1980 - 2015. (Unit: Bq m^{-3})

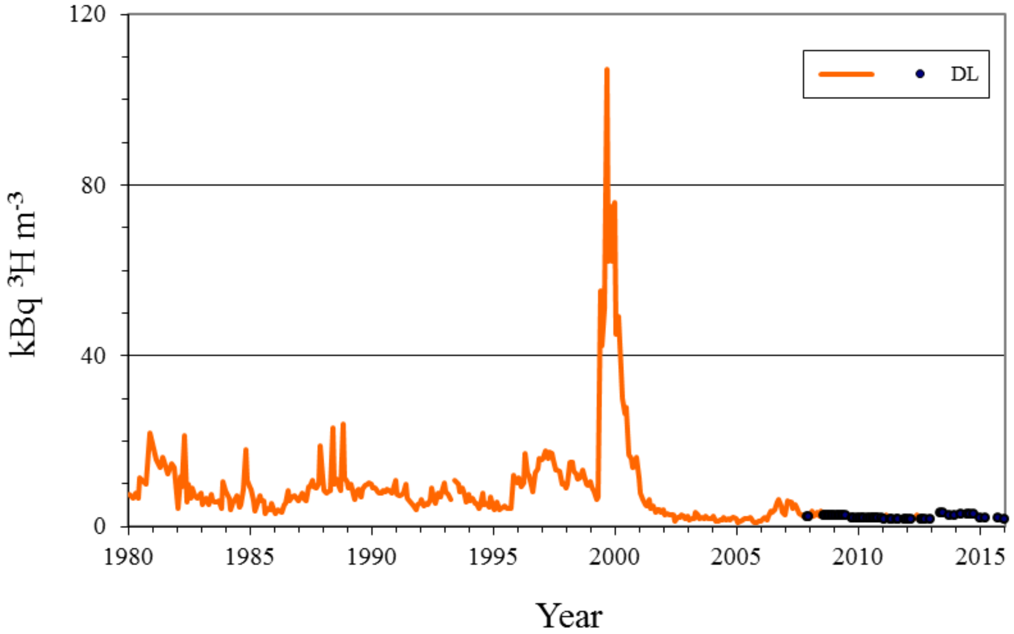

Fig. 4.2. Tritium in seawater collected in Roskilde Fjord 1980 - 2015. (Unit: kBq m^{-3}; DL = detection limit)

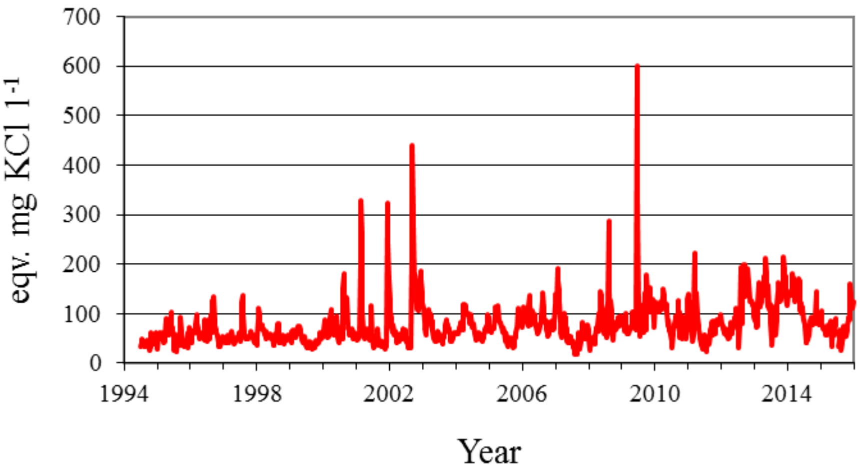

Fig. 7.1. Total-beta radioactivity in waste water collected at Risø 1994 - 2015. (Unit: eqv. mg KCl l^{-1})

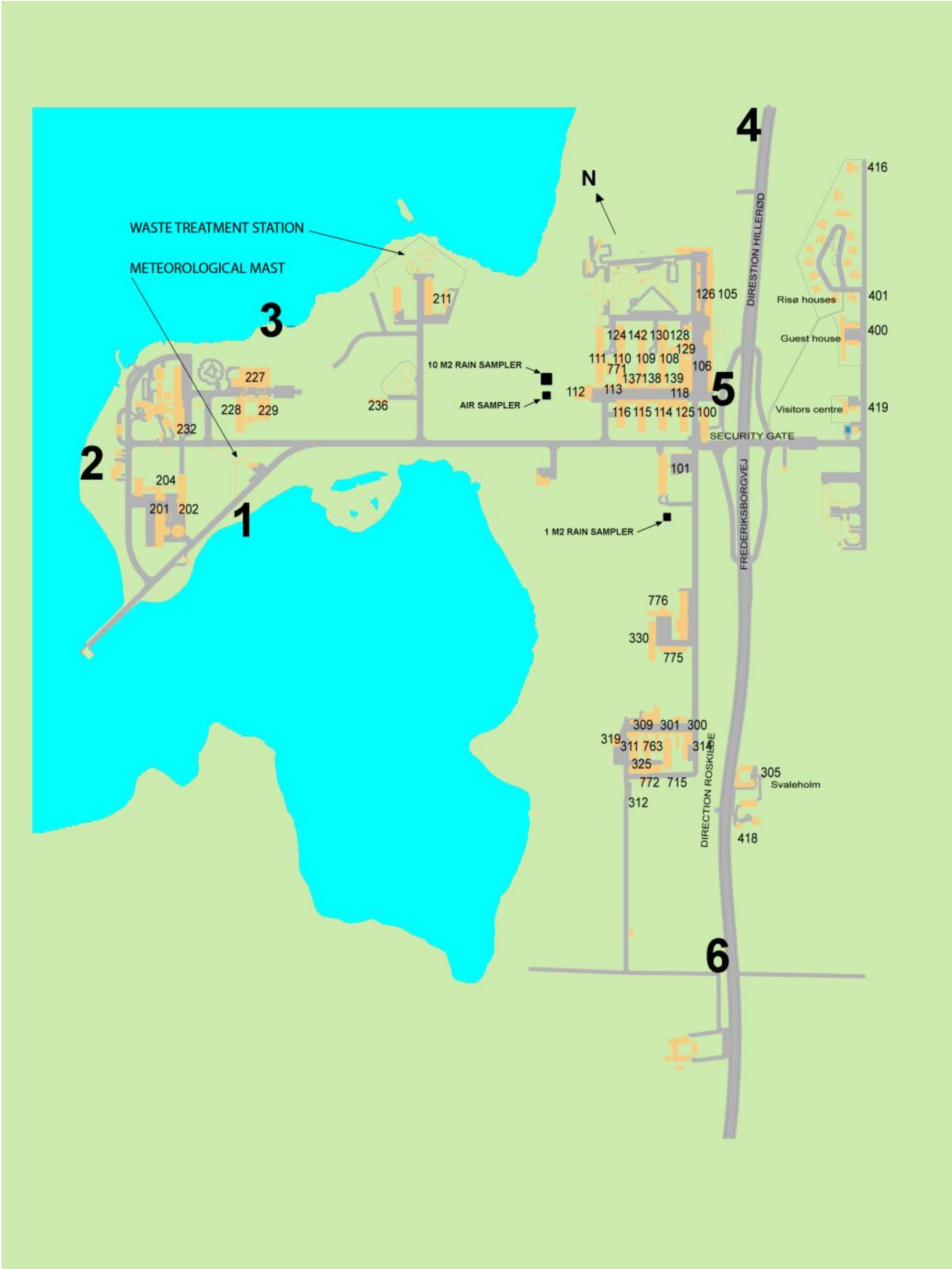

Fig. 8.1. Locations (1-6) for TLD measurements around the border of Risø (cf. Table 8.1).

Fig. 8.2. Locations for measurements of background radiation around Risø in Zones III, IV and V.

MATERIALS AND METHODS

External gamma dose rate monitoring

Monitoring of external gamma dose rate is carried out with the following devices

- Thermolumiscence dosimeters TLD: LiF, measurement frequency annually from May to April. TLD equipment manufacturer: ALNOR/RADOS
- NaI detector: 3x3 inch, SAM 935 Surveillance and Measurement System, Berkeley Nucleonics Cooperation, USA, visual read-out

Calibration of TLD is carried out by irradiation of dosimeters at a calibration irradiator. Traceability of delivered doses is ensured through calibration of the dose rate of the calibration irradiator by the National Institute of Radiation Protection (SIS). Calibration has been verified by measurement with ionisation chamber from NPL, UK. The NaI detector is calibrated periodically vs. a Reuter Stokes high-pressure ionisation chamber.

Air sampler

The sampler at Risø is manufactured by DTU. Air is drawn through a polypropylene filter at a rate of about 2000 m^3/h. The filter is normally changed weekly. The flow rate is monitored by a gas meter connected to a shunt. The gas meter reading is compared to that of a reference gas meter intermittently.

DTU analyse the filters by gamma spectrometry shortly after filter change to check for the presence of short-lived man-made radionuclides. The air filters are subsequently stored for a minimum of one week to allow for decay of short-lived naturally occurring radionuclides before repeated gamma analysis. Filters are analysed for ^{137}Cs, ^{7}Be and ^{210}Pb and other gamma emitters.

Deposition collector

The Risø site operates a large rain collector of 10 m^2. The collector is heated and water is passed through an ion exchange column to a large tank. The 10 m^2 collector provides monthly samples of rain water analysed for tritium and ion exchange resin which is analysed by gamma spectrometry for ^{7}Be, ^{137}Cs and ^{210}Pb and other gamma emitters.

Water and sediment

A waste water sample from the Waste Treatment Station is collected weekly and analysed for total beta radioactivity and the radionuclides ^{131}I, ^{137}Cs and ^{226}Ra. Water samples from Roskilde Fjord are collected each quarter and analysed for tritium, annually for ^{137}Cs. A sediment sample is collected annually from Roskilde Fjord and analysed for ^{137}Cs.

Terrestrial and aquatic biota and flora

Grass samples are collected weekly at the Risø site and analysed by gamma spectrometry. Samples are bulked to monthly samples which are analysed for ^{137}Cs.

Seaweed samples are collected annually from Roskilde Fjord at Risø and analysed for ^{137}Cs.

Sample reception and preparation

Sample identification numbers are entered in log books. Sample preparation methods include drying, freeze drying, ashing, sorting and sieving. Selected samples are archived.

Sample measurements

Radioactivity in samples is measured by total beta counting and gamma spectrometry.

Measurement devices

- Ge detectors for gamma spectrometry. Calibration of detectors is based on mixed-nuclide standards used occasionally. Monthly checks are made of detector efficiency and energy resolution. Background measurements of gamma systems are made a few times per year.
- Low-level Geiger-Müller counters for total beta counting, manufactured by DTU. Calibration based on standards of KCl. Counting efficiency and background are checked monthly.
- Liquid scintillation spectrometer for analysis of tritium in water. Samples are analysed with a calibration standard.

Analytical results, data handling and reporting tools

Analytical results are printed on paper, recorded in log books and stored in a data base on intranet. Results below detection limits recorded as such. Spreadsheets are used for calculating results from raw data.

Quality assurance, laboratory accreditation and intercomparison exercises

Analytical results are checked by experienced staff and discussed with senior scientists if questions arise.

DTU is accredited to testing for radioactivity by DANAK according to the international standard ISO 17025. The accreditation covers testing for certain non-gamma emitting radionuclides but not for radionuclides occurring in the environment and food in general.

DTU participate regularly in international intercomparisons on laboratory analyses of radionuclides.

CONCLUSIONS

This report shows the results of the environmental surveillance monitoring programme carried out at and around the Risø site in July-December 2015. The mean concentrations in air were: 0.24±0.18 μBq m^{-3} of ^{137}Cs, 2.71±0.88 mBq m^{-3} of ^{7}Be and 0.28±0.20 mBq m^{-3} of ^{210}Pb (±1 S.D.). The depositions by precipitation at Risø in the second half of 2015 were: 0.085±0.011 Bq m^{-2} of ^{137}Cs, 607±30 Bq m^{-2} of ^{7}Be, 59.6±4.2 Bq m^{-2} of ^{210}Pb and 1.4±0.2 kBq m^{-2} of ^{3}H. The average background dose rate (TLD) at Risø (Zone I) was measured as 42 nSv h^{-1} compared with 40 ± 2 nSv h^{-1} (±1 S.D.) in the four zones around Risø. None of the recorded levels of radioactivity and radiation have given rise to concern. A single sample (seawater) has not yet been measured for ^{137}Cs due to laboratory reconstruction work, but the result will be reported as soon as available.

Center for Nuclear Technologies is Denmark's national competency center for nuclear technology. With roots in research in the peaceful use of nuclear power, DTU Nutech works with the applications of ionizing radiation and radioactive substances for the benefit of society.

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